# Comparison of Several Detector Technologies for Measurement of Special Nuclear Materials[i]

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## Abstract

Gamma spectral analysis of measurements made on Special Nuclear Materials (SNM) provides identification and sometimes quantification of the SNM present. Different radiation detection technologies have been used for measurement of SNM. Selection of one of these for a particular application depends on the information that can be obtained using that detector and on the support needed to use the detection technology, especially if it is to be applied at sites where limited support is available. In some cases, detector selection may be determined by a need to limit the information available from measurements due to international treaty requirements. There are many studies of *system* capabilities in SNM measurement. Most of these used either scintillator or high-resolution (HPGe) detectors embedded in a unique electronics and software combination. Their respective developers claimed good results for their instrument. Unfortunately, the 'packaging' often obscured the question of what can be obtained from various detector technologies. While instrumentation and analysis is important, a study comparing *only* detector characteristics would serve as a starting point for development of systems for SMN measurement. Our detector study consisted of measurements on shielded and unshielded high-enriched uranium and Plutonium sources using detectors: NaI(Tl), Cadmium Zinc Telluride (CZT), Mercuric Iodide ($HgI_2$), and two HPGe detectors, one with liquid nitrogen cooling and one with electromechanical cooling. Measurements were made simultaneously using all detectors. As expected, all detectors are suitable for SNM measurements in cases where one radionuclide, such as $^{235}U$, is the dominant constituent, but only the high-resolution detectors are suitable for isotopic composition determinations using 'standard' spectroscopic analysis. Room temperature semiconductor detectors (CZT, $HgI_2$) can provide limited isotopic information. Development of improved analysis algorithms for room-temperature semiconductor detectors would be desirable because of their attractive characteristics: small sizes and minimal support requirements.

## Introduction

We undertook this task to provide an 'equal' comparison of radiation detector technologies for the measurement of SNM. There are (and have been) many studies of *system* capabilities in SNM measurement. Most of these used either scintillator or high-resolution (HPGe) detectors embedded in a unique electronics and software combination. Their respective developers claimed good results for their instrument. The 'packaging' often obscured the question of what can be obtained from various detector technologies. While instrumentation and analysis is important for this work, and user-friendly features are also important, a study comparing detector characteristics would serve as a starting point for the development of systems for arms control. Moreover, knowing the capabilities of various detectors would be valuable when we are the 'inspected' party rather than the 'inspector', required by various bilateral agreements.

The detector study consisted of measurements on shielded and unshielded SNM using detectors: NaI (Tl), Cadmium Zinc Telluride (CZT), Mercuric Iodide ($HgI_2$), and two HPGe detectors, one with liquid nitrogen cooling ("$LN_2$") and one with electromechanical cooling ("EMC"). A high pressure xenon ionization chamber was to be tested but did not arrive in time to be included.

## Gamma Spectroscopy of Special Nuclear Materials

The individual gamma ray energies used to establish the presence and quantify SNM are well established.[ii] These include the 143.7 and 185.6 keV gamma rays of $^{235}U$ and the 1001 keV emission of $^{234m}Pa$, a daughter of $^{238}U$. Plutonium gamma spectra are more complex: significant emission occurs at 59.5 keV from $^{241}Am$

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(daughter of 241Pu), 129.3 and 413.7 keV for $^{239}$Pu, 45.2 keV, and 160.3 keV for $^{240}$Pu. Both uranium and Plutonium emit many less intense gamma rays. Spectra of Plutonium mixtures are complex.

Measurement of $^{235}$U is more straightforward than measurement of the Plutonium isotopes. The gamma line at 185.7 keV is used most often. Unfortunately, in areas of high natural background radiation, the 186.2 and 187.1 keV gamma lines of $^{226}$Ra, a naturally occurring radionuclide, interfere with the 185.7 keV line of $^{235}$U. The three gamma lines can only be separated using high-resolution detectors under ideal measurement conditions. Use of the 143 keV gamma line of $^{235}$U is an alternative (the 143 keV line is used extensively in gamma spectroscopy in support of cleanup operations).

Practical measurements of Plutonium are more complex. There are five isotopes of interest: $^{238}$Pu, $^{240}$Pu, $^{239}$Pu, $^{241}$Pu, and $^{242}$Pu that are produced from irradiation of $^{238}$U. $^{239}$Pu is of most interest to weapons inspections. Due to spontaneous neutron emission, $^{238}$Pu and $^{240}$Pu are undesirable in weapons but are tolerable in Plutonium used in nuclear reactors. The ratio of $^{240}$Pu to $^{239}$Pu in 'weapons grade' materials is ~5%, while this ratio is ~15% for 'reactor grade' Plutonium. From an arms control viewpoint, the presence of any form of Plutonium may be significant, but only 'weapons grade' material is typically used in nuclear weapons. Fortunately, the individual isotopes of interest emit gamma rays having unique energies; unfortunately the spectral are complex and require high-resolution measurements to separate the individual gamma lines. Codes have been developed for the determination of SNM content based on spectra. For example, the MGA code[iii], available in several different forms, is often used for determining the isotopic composition of Plutonium samples.

Additionally, some gamma rays from the daughter isotopes of SNM are useful in identification of SNM by association. For example, $^{232}$U has been suggested as a means to identify the presence of $^{235}$U, using the 2614 keV gamma line of $^{208}$Tl, a daughter of $^{232}$U.[iv]. $^{232}$U is a small constituent of 'high enriched uranium' (HEU). $^{208}$Tl produces a 2614 keV gamma ray, however, $^{208}$Tl is also produced from the natural $^{232}$Th decay chain. Using the 2614 keV (or 583 keV) gamma line as an indication of $^{235}$U requires a correction for the $^{208}$Tl produced by natural sources. Measurement of the 59.5 keV gamma ray emitted by $^{241}$Am (a daughter of $^{241}$Pu) is often used as an indication of Plutonium; the 59.5 keV gamma has been used as a measure of Plutonium contamination in environmental surveys.[v] $^{241}$Am concentration for Plutonium samples may be very different. Plutonium samples used for this study both emit gamma rays at 59.5 keV, with the 'weapons grade' sample having much higher emission. This difference is not a characteristic of 'reactor' and 'weapons grade' Plutonium, but an indication of the time that has elapsed since the Plutonium sample was separated from neutron-irradiated Uranium (i.e., U-238).

Lastly, the gamma ray background in the facility in which the inspector performs his measurements must to be considered. Background contributions to the measured spectrum determine the minimum detectable activity in spectral measurements. For identification purposes, extra gamma ray lines complicate the analysis. The facility background in which these measurements were done contains contributions due to $^{137}$Cs, $^{40}$K, $^{228}$Ac, $^{214}$Bi, $^{226}$Ra, and $^{212}$Pb.

**Intercomparison Measurements**

Detectors based on several different technologies were selected for comparison. These included high resolution HPGe: a 55% efficiency detector with EMC, a 40% efficient HPGe with $LN_2$ cooling, a 10 mm x 10 mm x 7.5 mm coplanar grid CZT detector, a 10 mm x 10mm x 2.8 mm $HgI_2$detector, and a 2" diameter x 2" long NaI (Tl) detector with a photomultiplier. These detectors represent a cross-section of available technology. To aid in comparisons of detectors of varying sizes, we experimentally determined the efficiency vs. energy for each detector and calculated the 'area-normalized efficiencies': ratios of 'photopeak count rate / gamma rays striking the detector' measured at multiple specific energies. Figure 1 shows the

area-normalized efficiencies vs. energy for our detector group. Absolute efficiencies can be estimated by multiplying the area-normalized efficiency by the source solid angle fraction viewed by the detector.

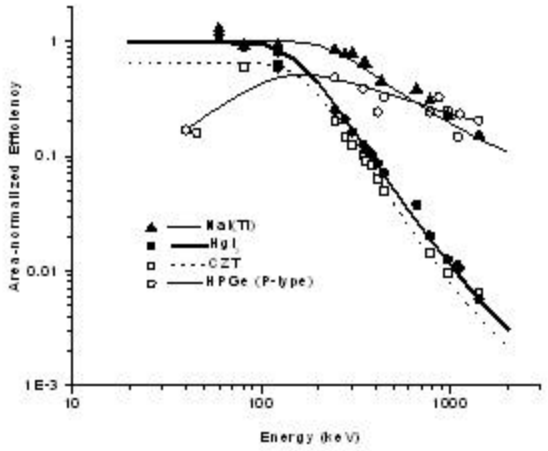


Figure 1 Area-normalized Detector Efficiencies

Information shown in Figure 1 includes the efficiency vs. energy trends for the CZT and $HgI_2$ detectors – a more rapid decrease in efficiency than the others due to smaller detector active thicknesses. The CZT detector efficiency never reaches unity; counts are always lost even at low energies. The HPGe efficiency is representative of both the liquid nitrogen and electromechanically cooled P-type detectors. HPGe and NaI(Tl) detector efficiency curves have shapes similar to those reported previously.

The detectors were arranged in a "circle" surrounding a test source in the TA-18 facility at Los Alamos National Laboratory (LANL). Detectors and sources rested on a wooden floor built over a concrete basement. Metal walls and roof covered the wooden floor. Locations of the various detectors were selected to provide a "good" count rate without significant dead time. Figure 2 shows the arrangement. This geometry was maintained during SNM measurements. Counting (live) times were 20 minutes for the $LN_2$ and EMC detectors and 60 minutes for the others. All detectors were "aimed" at the source to achieve the highest possible count rates.

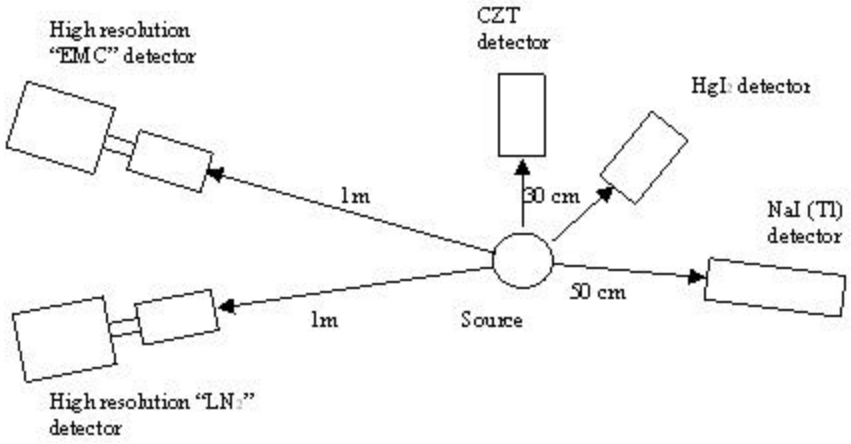


Figure 2 Measurement Arrangement

The LANL NIS-6 group provided small cylindrical SNM samples of four isotopic types: 1)'HEU' (93.15 % $^{235}$U, remainder $^{238}$U), 2) depleted uranium (0.22 % $^{235}$U by weight), 3) weapons grade Plutonium (6% $^{240}$Pu by weight, assumed 94% $^{239}$Pu), and 4) 'high burnup' Plutonium (21.94% $^{240}$Pu by weight, assumed 78% $^{239}$Pu). Sources from the four isotopic types in both shielded and unshielded configurations were measured individually to generate the spectra shown in this paper.

**SMN Measurement Results**

All detectors were tested using several HEU samples, both shielded and unshielded. Comparisons of the detectors were made based on good resolution of the 143, 160, and 185 keV gamma lines needed to positively identify HEU. The 185 keV gamma line is often used alone for "enrichment" measurements, but use of the other lines is needed when little is known about the sample in advance. Detector comparisons are shown below. Comparisons of shielded and unshielded sources are made in the next section.

The first spectrum (Figure 3) shows data from a 295 g unshielded HEU sample with a spectrum of facility background, both measured with the EMC detector. Counting times were 20 minutes for the sample and overnight for the background. Data using the $LN_2$ detector were identical to those shown for the EMC. The majority of gamma lines visible are due to natural background radiation, including $^{137}$Cs (661 keV), $^{40}$K (1460 keV) and $^{214}$Bi (1764 keV). This spectrum illustrates the complexity of spectra measured in a legacy nuclear facility with considerable industrial use.

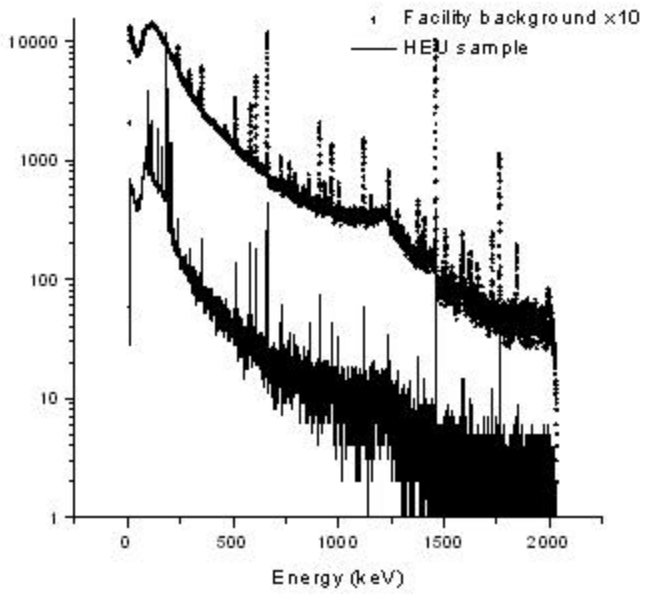


Figure 3 Facility Background and HEU Sample High-resolution spectra

Note the abundance of gamma lines from the background. Only a few low energy lines due to $^{235}$U are visible. Ideally, the sample-detector geometry can be adapted to minimize the effects of natural background.

Figure 4 shows HEU spectra from the four detectors. As expected, the HPGe detectors resolved the 143, 160, 185, and 205 keV line grouping and the uranium x-rays around 95 keV. The room-temperature semiconductor detectors (CZT and $HgI_2$) resolved the four gamma lines but not the x-rays and the NaI(Tl) detector only resolved the 185 keV gamma line. Count rates in this and many figures have been scaled to show several spectra on a single plot. Peak heights may not represent actual count rates measured during this work.

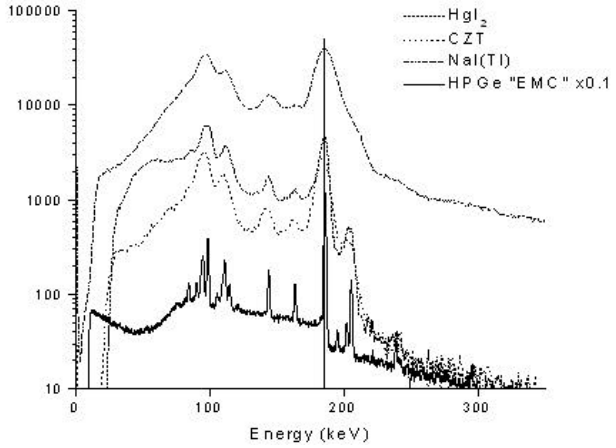


Figure 4 HEU Spectra Obtained Using Several Detectors. The 185 keV Peak is Marked.

We obtained spectra of measurements of weapons grade Plutonium. Figures 5-6 show these results.

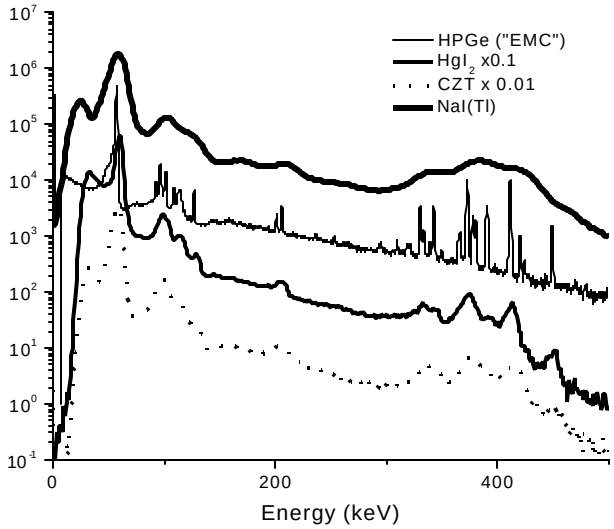


Figure 5 Weapons Grade Plutonium Detector Comparison, Low-energy Region

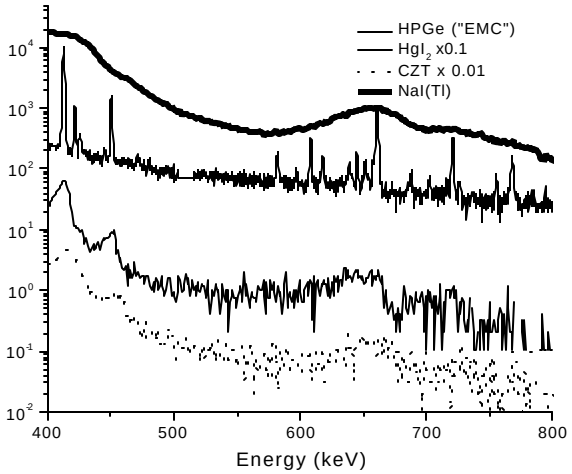


Figure 6 Weapons Grade Plutonium detector Comparison, High-energy Region

Figures 5 and 6 show that all the detectors are suitable for semi- quantitative measurements based on the 414 keV region, but the higher resolution spectra are required for isotopic analysis of Plutonium samples. CZT and mercuric iodide spectra can be used for isotopic analysis if state-of-the-art computer codes are available. Spectra acquired with a 'best of the best' CZT detector were used as input to the MGA code for determining Plutonium isotopics.[vi] In the high energy region, however, only the spectra acquired using the HPGe detector shows significant detail.

Differentiation between weapons grade and high-burnup Plutonium is important because high burnup Plutonium is typically not used in weapons. We compare detector performance in measuring both weapons grade and high burnup Plutonium in Figures 7, 8, and 9.

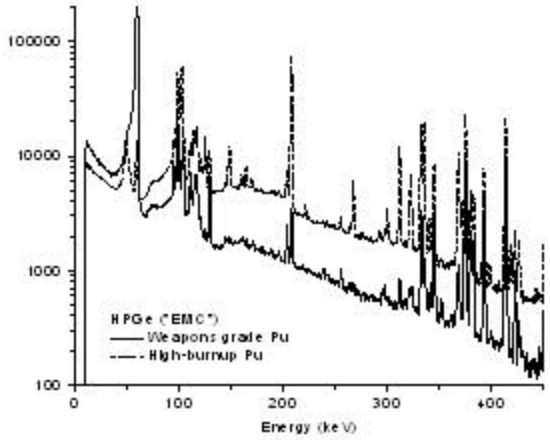


Figure 7 High Resolution Spectra of Weapons Grade and High Burnup Plutonium, Low-energy Region

Numerous examples of Figure 7 can be found in published data. For comparison, Figure 8 shows spectra from measurement of the same samples using the CZT detector, and Figure 9 shows spectra of the high energy region using the CZT detector. Spectra acquired using the mercuric iodide detector closely resemble their CZT counterparts. Low energy (50-450 keV) data from the CZT or mercuric iodide detectors could be useful in differentiating between weapons grade and high burnup Plutonium, perhaps by using spectral peak ratios, with some difficulty. Data above 450 keV is degraded due to the decreasing detector efficiency at higher energies. Data acquired using the NaI(Tl) detector did not show significant differences. The 59.5 keV peak from $^{241}$Am, typically present in weapons grade Plutonium, may not be present in more recently processed material or material that did not originally have much $^{241}$Pu.

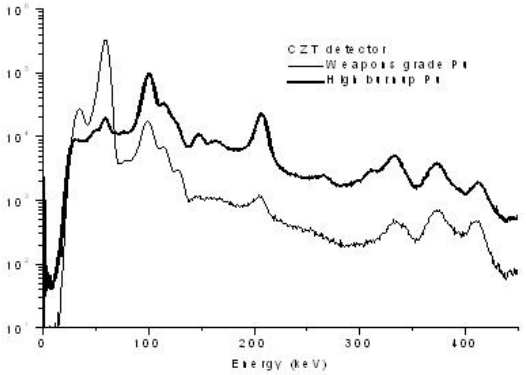


Figure 8 Weapons Grade and High Burnup Plutonium, 0-450 keV

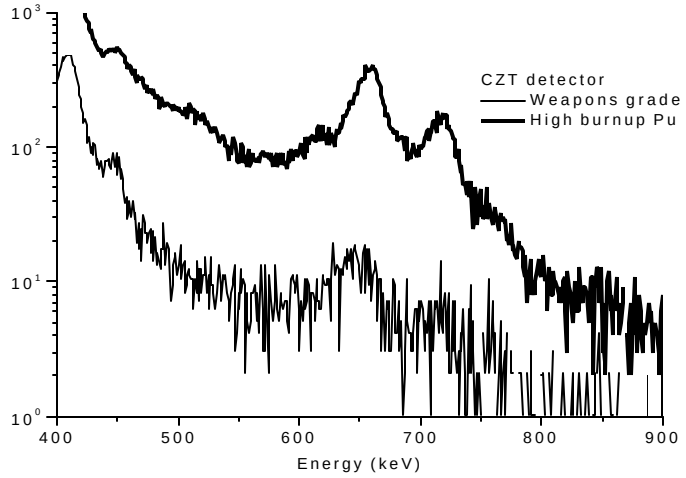


Figure 9 Weapons Grade and High Burnup Plutonium, 400-900 keV

We also investigated the effects of shielding on spectra taken with the different detectors. Shielding in our tests consisted of 1" of steel added between the source and detector. While actual shielding of SNM covers an unpredictably wide range, we sought to determine the effects of shielding on recognizable peak shapes that would be used as input to radionuclide identification algorithms. We did not attempt to determine quantitative results. Figure 10 shows the effects of shielding on HEU spectra acquired using HPGe and $HgI_2$ detectors. The 185 keV peak is attenuated but still present and the higher energy peaks are less attenuated. The uranium x-rays are highly attenuated, thereby complicating the process of differentiating uranium from natural radium, for example. Similar effects can be observed in spectra of shielded and unshielded Plutonium samples.

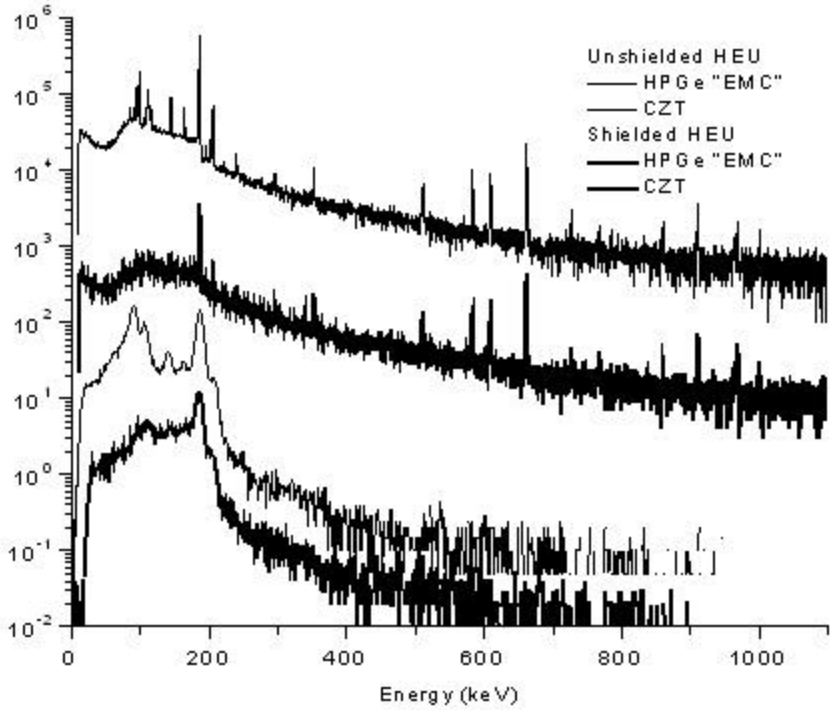


Figure 10 Spectra from Shielded and Unshielded HEU Samples (scaled to separate spectra)

Examination of the spectra show that a typical peak search algorithm would 'find' the $^{235}U$ peaks in all four spectra, although peaks are clearly better defined in the HPGe data. Again spectra acquired using mercuric iodide and CZT detectors are almost identical in terms of peak resolutions and relative peak heights. HPGe and CZT detectors have been successfully implemented in radionuclide identification devices.

**HPGe Detector Comparison**

High-resolution spectra shown in previous figures were obtained using Constellation Technology's EMC HPGe detector. The EMC HPGe detector was developed for arms control inspectors and others who may not have access to liquid nitrogen at the measurement locations. In all measurements, the EMC HPGe detector and the conventional liquid nitrogen-cooled detectors performed identically. This is shown in Figure 11, below, showing a portion of the weapons grade Plutonium spectrum.

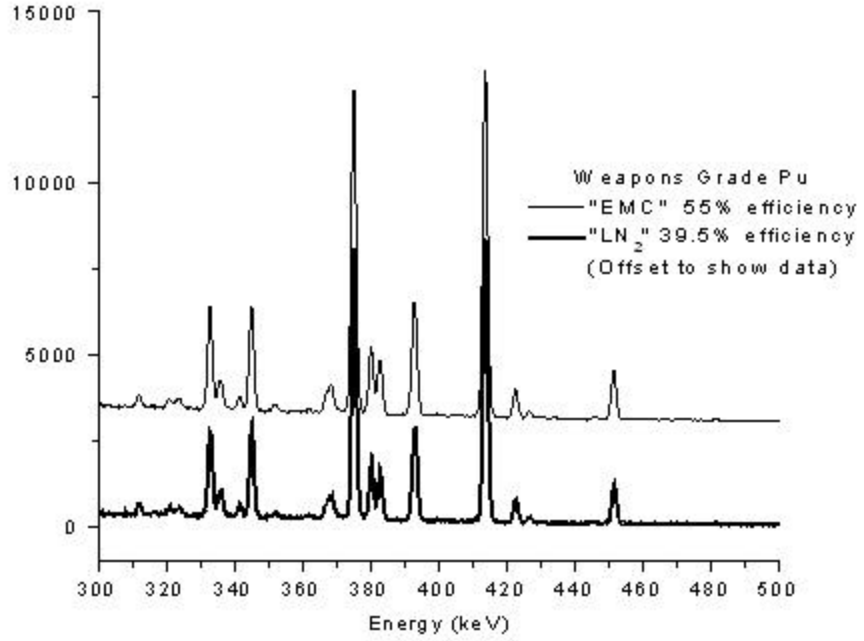


Figure 11 HPGe Detector Comparison

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[i] This work was supported by the Defense Threat Reduction Agency

[ii] For example, see: D. Reilly, et. al *Passive Nondestructive Assay of Nuclear Materials*. NUREG/CR-5550 (1991), p 18.

[iii] R. Gunnink, W. Ruhter. :MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances", vols 1-3, UCRL-LR-103220 (1990).

[iv] A. Perrung, Predicting U-232 Content in Uranium, PNNL-12075 (1998)

[v] See for example, A.Proctor, Aerial Radiological Surveys, DOE/NV/11718-127 (1997).

[vi] R. Gunnink, private communication to A. Proctor ( 2002)